

## **CERTIFICATION**

I, Shigeru Yanagihara, of Yanagihara & Associates, 503, Nishishinbashi Chuo Bldg., 15-8, Nishishinbashi 3-Chome, Minato-Ku, Tokyo 105-0003, Japan, hereby certify that the following is a true and correct translation, to the best of my knowledge and belief, of an official certified copy of the document filed in respect of Japanese Patent Application No. 107866/1997 on 24th April 1997.

Place

Tokyo

Date

November 21, 2003

Shigeru YANAGIHARA

## [ DESIGNATION OF THE DOCUMENT ]

#### **SPECIFICATION**

### [ TITLE OF THE INVENTION]

RESIN COMPOSITION BASED ON CRYSTALLINE POLYPROPYLENE

### [ PATENT CLAIMS ]

[Claim 1] A resin composition based on crystalline polypropylene, comprising

- (a) 20 35 % by weight of a component soluble in paraxylene of  $23^{\circ}$ ,
- (b) 43 65 % by weight of a component soluble in paraxylene of 135  $^{\circ}{\rm C}$  and insoluble in paraxylene of 23  $^{\circ}{\rm C}$  and

#### wherein

the component (a) soluble in paraxylene of 23  $^{\circ}$ C is composed substantially of an elastomer (A) having a content of styrene or its derivative in the range of 0 - 35  $^{\circ}$ 6 by weight and an intrinsic viscosity [ $\eta$ ] determined in decalin at 135 $^{\circ}$ C in the range of 0.1 - 5 d1/g,

the component (b) soluble in paraxylene of 135℃ and insoluble in paraxylene of 23 ℃ is composed substantially of a crystalline polypropylene (B) having an isotactic pentad proportion (mmmm) of 98 % or higher, a molecular weight distribution expressed by Mw/Mn, determined by gel permeation chromatography, of 9 or higher and a molecular weight distribution expressed by Mz/Mw of 8 or higher and

the component (c) insoluble in paraxylene of 135°C is composed substantially of talc (C) having an average particle size in the range of 1 - 5  $\mu$  m.

- [Claim 2] A resin composition based on crystalline polypropylene as claimed in Claim 1, wherein the elastomer (A) consists of an elastomer based on styrene consisting of
  - (A-1) an elastomer based on styrene composed of 10 - 40 % by weight of a constituent polymer block based on styrene and 60 - 90 % by weight of a constituent polymer block based on a conjugated diene;
  - (A-2) an ethylene/ $\alpha$ -olefin random copolymer; or
  - (A-3) an ethylene/ $\alpha$ -olefin/non-conjugated polyene random copolymer.

[Claim 3] A resin composition based on crystalline polypropylene as claimed in Claim 1 or 2, wherein the crystalline polypropylene (B) consists of a crystalline polypropylene having an ethylene content of 0-3% by weight and a melt flow rate, determined at 230 °C under a load of 2160 g, of 30 - 150 g/10 min.

# [ DETAILED DESCRIPTION OF THE INVENTION ]

[ 0001]

## [ TECHNICAL FIELD OF THE INVENTION]

The present invention relates to resin composition crystalline polypropylene, based on specifically, to а resin composition based on crystalline polypropylene used favorably for exterior articles of automobile.

[ 0002]

#### [ PRIOR ART ]

For exterior articles and parts for automobile made of moldings of polypropylene resin composition, a feeling integrated with car body, a superior appearance in favor of providing a high grade and high designing impression and requisite strength etc. are required. Also for parts of domestic electrified appliances, high grade feeling, mechanical strength and so requested. Heretofore, there have been in practical application, in the application field requiring superior appearance, mechanical strength and many resin compositions based on polypropylene, in which olefinic elastomers, talc, pigments and so on, are blended together with polypropylene.

[ 0003]

When, however, such a polypropylene-based resin composition as above is molded by injection molding, a deterioration in the appearance of the molded products may, in some cases, occur due to occurrence of "flow marks", namely, stripes caused by repetition of high- and low-gloss regions on the surface of the molding.

[ 0004]

## [ SUBJECT TO BE SOLVED BY THE INVENTION ]

The subject of the present invention is to provide composition a resin based on crystalline polypropylene which can bring about molded products without suffering from occurrence of flow-mark and. exhibiting therefore, a superior appearance excellent mechanical properties, such as mechanical strength and so on.

[ 0005]

### [ MEANS FOR SOLVING THE SUBJECT ]

The present invention relates to a resin composition based on crystalline polypropylene which is to be defined as follows:

- (1) A resin composition based on crystalline polypropylene, comprising
- (a) 20 35 % by weight of a component soluble in paraxylene of  $23^{\circ}$ ,
- (b) 43 65 % by weight of a component soluble in paraxylene of 135  $^{\circ}{\rm C}$  and insoluble in paraxylene of 23  $^{\circ}{\rm C}$  and
- (c) 15 22 % by weight of a component insoluble in paraxylene of 135  $^{\circ}$  ,

#### wherein

the component (a) soluble in paraxylene of 23  $^{\circ}$ C is composed substantially of an elastomer (A) having a content of styrene or its derivative in the range of 0 - 35  $^{\circ}$ 6 by weight and an intrinsic viscosity [ $\eta$ ] determined in decalin at 135 $^{\circ}$ C in the range of 0.1 - 5 dl/g,

the component (b) soluble in paraxylene of 135°C and insoluble in paraxylene of 23 °C is composed substantially of a crystalline polypropylene (B) having an isotactic pentad proportion (mmmm) of 98 % or higher, a molecular weight distribution expressed by Mw/Mn, determined by gel permeation chromatography, of 9 or higher and a molecular weight distribution expressed by Mz/Mw of 8 or higher and

the component (c) insoluble in paraxylene of 135°C is composed substantially of talc (C) having an average particle size in the range of 1 - 5  $\mu$  m.

- (2) A resin composition based on crystalline polypropylene as defined in the above (1), wherein the elastomer (A) consists of an elastomer based on styrene consisting of
  - (A-1) an elastomer based on styrene composed of 10 - 40 % by weight of a constituent polymer block based on styrene and 60 - 90 % by weight of a constituent polymer block based on a conjugated diene;
  - (A-2) an ethylene/ $\alpha$ -olefin random copolymer; or
  - (A-3) an ethylene/ $\alpha$ -olefin/non-conjugated polyene random copolymer.
- (3) Α resin composition based on crystalline polypropylene as defined in the above (1) or (2), wherein the crystalline polypropylene (B) consists of a crystalline polypropylene having an ethylene content of 0 - 3 % by weight and a melt flow rate, determined at 230 °C under a load of 2160 g, of 30 - 150 g/10 min.

[ 0006]

The component (a) soluble in paraxylene of 23  $^{\circ}\mathrm{C}$ according to the present invention is composed substantially of an elastomer (A) having a content of styrene its derivative (designated hereinafter sometimes simply as the styrene content) in the range of 0 -35 % by weight, preferably 0 - 30 % by weight, and an intrinsic viscosity determined in decalin at 135  $^{\circ}$ in the range of  $0.1 - 5 \, dl/g$ , preferably  $0.3 - 3 \, dl/g$ .

Thus, the component (a) is composed substantially of the elastomer (A) and preferably constituted entirely of the elastomer (A), while it is permissible that a small amount, for example, not higher than 10 % by weight, preferably not higher than 5 % by weight, of other constituents, such as an atactic polypropylene having low crystallinity and so on, may be contained.

[ 0007]

The elastomer (A) may be composed of one single a mixture of two or more elasomers or a elastomer, mixture of one or more elastomers with minor proportion of other ingredient(s). In case it is a mixture, the styrene content and the intrinsic viscosity mentioned above should refer to the average value for constituent ingredients of the mixture, wherein it is permissible that an elastomer having a styrene content and/or an intrinsic viscosity out of the above identified range may be included.

[8000]

Concrete examples of the elastomer (A) include elastomers based on styrene (A-1); random copolymers of ethylene/ $\alpha$ -olefin (A-2); random copolymers of ethylene/ $\alpha$ -olefin/non-conjugated polyene (A-3); other elastomeric polymers; and mixtures of them.

[ 0009]

As the styrene-based elastomer (A-1), there may be exemplified block-copolymers based on · styrene of a constituent polymer block based styrene (denoted hereinafter sometimes as the styrenic block) and a constituent polymer block based on conjugated diene (denoted hereinafter sometimes as the dienic block); styrene/butadiene random copolymers; styrene/isoprene random copolymers; and styrene/chloroprene random copolymers as well as hydrogenation products of them. Among them, preference is given to block-copolymers based on styrene.

The content of styrene or its derivative in the styrene-based elastomer (A-1) may be 5-35% by weight, preferable 10-30% by weight.

[ 0010]

The constituent polymer block based on styrene constituting the styrene-based block-copolymers constituted of styrene or a derivative thereof, wherein the concrete monomer thereof includes styrene, methylstyrene, p-methylstyrene, chlorostyrene and vinylnaphthalene. Among them, preference is given to These monomers may be used either alone or in styrene. combination of two or more of them.

Concrete examples of the monomers constituting the polymer block based on conjugated diene include butadiene, isoprene and chloroprene. Among them, butadiene and isoprene are preferred. These monomers may be used either alone or in combination of two or more of them.

[ 0011]

There is no special limitation in the form of binding between the styrenic block and the dienic block in the block-copolymers based on styrene, while a binding form of styrenic block-dienic block or of styrenic block-(dienic block-styrenic block), in which

n stands for a number of 1 - 5, is preferred.

The styrene-based block-copolymer can be produced by, for example, carrying out a block-copolymerization in an inert solvent in the presence of a lithium catalyst or a Ziegler catalyst. Details of the production procedures are given in, for example, Japanese Patent Publication Sho-40-23798 B.

[ 0012]

As the styrene-based block-copolymer, those of the hydrogenation products are preferred, while those which are not subjected to hydrogenation may also be employed. On the hydrogenation, the double bonds in the dienic blocks are hydrogenated, wherein the yield of hydrogenation may favorably amount to 90 mole % or more, preferably 97 mole % or more, of the total dienic blocks.

The hydrogenation treatment can be effected in an inert solvent in the presence of a known hydrogenation catalyst. Details of the procedures are given in, for example, Japanese Patent Publications Sho-42-8704 B, Sho-43-6636 B and Sho-46-20814 B.

[ 0013]

The block-copolymers based on styrene may favorably have a melt flow rate (MFR), determined at  $230^{\circ}$ C under a load of 2160 g, of 0.1 g/10 min. or higher, preferably 0.3 - 20 g/10 min.

[ 0014]

Concrete examples of the block-copolymers based on styrene include styrene/ethylene/butene/styrene block-copolymer (SEBS), styrene/ethylene/propylene/

styrene block-copolymer (SEPS), styrene/butadiene/
styrene block-copolymer (SBS), styrene/isoprene/styrene
block-copolymer (SIS) and styrene/ethylene/propylene
block-copolymer (SEP) as well as hydrogenation products
of them.

For the styrene-based block-copolymer, commercial products can be employed. Concrete examples include SEPTON (trademark, a product of Kuraray Co., Ltd.), KRATON G (trademark, a product of Shell Kagaku K.K.) and TUFTEC (trademark, a product of Asahi Chemical Ind. Co., Ltd.).

[ 0015]

The ethylene/ $\alpha$ -olefin random copolymer (A-2) is a random copolymer rubber made of ethylene and an  $\alpha$ -olefin having 3 - 20 carbon atoms.

As the  $\alpha$ -olefin having 3 - 20 carbon atoms, there may be exemplified concretely propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene and 1-eicosene. These  $\alpha$ -olefins may be used either solely or in combination of two or more of them. Among them, preference is given especially to propylene, 1-butene, 1-hexene and 1-octene.

[ 0016]

The ethylene/ $\alpha$ -olefin random copolymer (A-2) may favorably have an ethylene/ $\alpha$ -olefin mole ratio in the range of 95/5 - 70/30, preferably 90/10 - 75/25. In the copolymer, styrene or its derivative may be co-polymerized in a small proportion, but preferably without being co-polymerized.

The ethylene/ $\alpha$ -olefin random copolymer (A-2) may favorably have an MFR, at 230  $^{\circ}$ C under a load of 2160 g, of 0.1 g/10 min. or higher, preferably 0.5 - 5 g/10 min.

[ 0017]

The ethylene/ $\alpha$ -olefin/non-conjugated polyene random copolymer (A-3) is a random copolymer rubber made of ethylene, an  $\alpha$ -olefin having 3 - 20 carbon atoms and a non-conjugated polyene. As the  $\alpha$ -olefin having 3 - 20 carbon atoms, the same monomers as above are enumerated.

the non-conjugated polyene, there may exemplified acyclic dienes, such as 5-ethylidene-2norbornene, 5-propylidene-5-norbornene, dicyclopentadiene, 5-vinyl-2-norbornene, 5-methylene-2-norbornene, 5-isopropylidene-2-norbornene and norbornadiene; chainformed non-conjugated dienes, such as 1,4-hexadiene, 4-methyl-1,4-hexadiene, 5-methyl-1,4-hexadiene, methyl-1,5-heptadiene, 6-methyl-1,5-heptadiene, methyl-1,7-octadiene and 7-methyl-1,6-octadiene; trienes, such as 2,3-diisopropylidene-5-norbornene. Among them, 1,4-hexadiene, dicyclopentadiene and 5ethylidene-2-norbornene are used favorably.

[ 0018]

The ethylene/ $\alpha$ -olefin/non-conjugated polyene random copolymer (A-3) may favorably have a mole ratio of ethylene/ $\alpha$ -olefin/non-conjugated polyene in the range of 90/5/5 - 30/45/25, preferably 80/10/10 - 40/40/20. It is permissible that small proportion of styrene or a derivative thereof may be co-polymerized,

but preferably not be co-polymerized.

The ethylene/ $\alpha$ -olefin/non-conjugated polyene random copolymer (A-3) may favorably have an MFR, at 230 °C and 2160 g load, of 0.05 g/10 min. or higher, preferably 0.1 - 10 g/10 min.

Concrete examples of the ethylene/ $\alpha$ -olefin/non-conjugated polyene random copolymer (A-3) include ethylene/propylene/diene ternary copolymers (EPDM) and so on.

[ 0019]

According present to the invention, the component (.b) soluble in paraxylene of 135℃ insoluble in paraxylene of 23 ℃ is costituted substantially of a crystalline polypropylene (B) having an isotactic pentad proportion (mmmm) of 98 % or more, preferably 98.5 or molecular more, a weight distribution in terms of weight-average molecular weight/number-average molecular weight (Mw/Mn), determined by gel permeation chromatography (GPC), of 9 or higher, preferably 11 or higher, and a molecular weight distribution expressed by z-average molecular weight/weight-average molecular weight (Mz/Mw) of 8 or higher, preferably 9 or higher. The component (b) is constituted substantially of a crystalline polypropylene (B), preferably entirely of crystalline polypropylene (B), wherein it is permissible that other ingredient(s) may be contained in a small proportion, for example, 10 % by weight or less, preferably 5 % by weight or less.

[ 0020]

The isotactic pentad proportion (mmmm) refers

to a proportion of isotactic chains in terms of pentad chain of the molecular the crystalline polypropylene (B) determined by '3C NMR, namely, the proportion of number of propylene monomer units present each in the center of a chain of 5 successive propylene monomer units bound with each other by a meso-linkage relative to the total monomer units. Concretely, it is determined by the per cent fraction of the mmmm peak relative to the total absorption peaks in the methyl carbon region in a 13 C NMR spectrum.

[ 0021]

If the values of Mw/Mn and Mz/Mw are in the above-identified ranges, a small amount, for example, 0.5-20 % by weight, of polypropylene of an ultra high molecular weight of, for example,  $1 \times 10^6 - 1 \times 10^7$ , may be contained therein.

[ 0022]

crystalline polypropylene (B) composed of a crystalline polypropylene of species or of a mixture of two or more species of crystalline polypropylene or, further, a mixture of at least crystalline polypropylene with small proportion of other ingredient(s). In the case mixture, the isotactic pentad proportion (mmmm), Mw/Mn and Mz/Mw refer each to the average value with respect to the mixture and it is permissible polypropylene(s) having isotactic pentad proportion (mmmm), Mw/Mn and/or Mz/Mw out of the above-identified range and/or other resin(s) may be included.

[ 0023]

The crystalline polypropylene (B) may be constituted of a homopolymer of propylene or a copolymer with a small proportion, for example, 3 % by weight or less, of other  $\alpha$ -olefin(s) or so on. Concretely, it may contain, for example, 0 - 5 % by weight, preferably 0 - 3 % by weight, of ethylene.

The crystalline polypropylene (B) may favorably have a melt flow rate (MFR), determined at 230% under a load of 2160 g, of 30 - 150 g/10 min., preferably 30 - 100 g/10 min.

[ 0024]

The crystalline polypropylene (B) can be produced by known technique in the presence of a Ziegler-Natta catalyst composed of titanium trichloride and an alkyl aluminum compound or a composite catalyst composed of a magnesium compound and a titanium compound.

[ 0025]

According to the present invention, the component (c) insoluble in paraxylene of 135 °C is constituted of talc (C) having an average particle size of 1 - 5  $\mu$  m, preferably 1 - 3  $\mu$  m.

[ 0026]

The content of each component of the composition according to the present invention may be 20 - 35 % by weight, preferably 22 - 30 % by weight, for the component (a), 43 - 65 % by weight, preferably 50 - 62 % by weight, for the component (b) and 15 - 22 % by weight, preferably 16 - 20 % by weight, for the component (c).

the composition according to the present invention, the component (a) is composed substantially of the elastomer (A) and constituent(s) soluble in 23 ℃ other than the elastomer (A) scarcely be included or even entirely excluded. The (b) is composed substantially component the crystalline polypropylene (B) and constituent(s) in 135℃ soluble paraxylene and insoluble in 23 ℃ paraxylene other than the crystalline polypropylene (B) may only scarcely be included or even entirely excluded. The component (c) is composed substantially of the talc (C) and constituent(s) insoluble in 135 ℃ paraxylene other than the talc (C) may only scarcely be included or even entirely excluded.

[ 0027]

The composition according to the present invention can be obtained by blending or melt-kneading the polymers to be blended and the talc hacing an average particle size of 1 - 5  $\mu$  m, preferably 1 - 3  $\mu$  m, on a mixing means, such as Bumbury's mixer, monoor bi-axial extruder or high-speed biaxial extruder, in such a mixing ratio that the resulting mixture will have contents of the components (a) to (c) each in the range given above and that the components (a), (b) and (c) will be composed substantially of the elastomer (A), the crystalline polypropylene (B) and the talc (C), respectively.

[ 0028]

As the polymers to be blended upon the preparation of the composition according to the present

to be served in the invention, polymers prepared composition as the elastomer (A) or as the crystalline polypropylene (B) may be used without any restriction. Concretely, there may be enumarated therefor, addition to the elastomer (A) and the crystalline polypropylene (B) mentioned above, crystalline polypropylenes (B) containing the elastomer (A), example, the crystalline polypropylene (B) containing the elastomer (A) by-produced upon the production of the crystalline polypropylene (B).

[ 0029]

The composition according to the present invention obtained in the manner as above is superior in the mechanical properties, such as strength and so on and can be molded by, for example, injection molding and so on, into articles exhibiting superior appearances without suffering from occurrence of flow marks.

[ 0030]

the composition according to the In invention, other additives which have conventionally been used in resin compositions based on polyolefin, example, heat stabilizer, antistatic weathering stabilizer, light stabilizer, antiaging agent, antioxidant, softener, dispersant, coloring agent and lubricant, may be blended in amounts not obstructing the purpose of the present invention.

[ 0031]

The composition according to the present invention can be used as the material in the field in

which polypropylene has found its use, for example, for interior and exterior parts for automobile, housings of domestic electric appliances, office supplies, household articles and miscellaneous articles, wherein it is particularly preferably used for the raw material for automobile exterior articles. Concrete examples of automobile exterior articles include bumper, side mole, garnish, window mole and wheel housing cover. Among them, use for the raw material of bumper is favorable.

[ 0032]

The articles made from the composition according to the present invention, such as exterior articles of automobile and the like, have superior mechanical properties and are also superior in the appearance without exhibiting flow mark. For molding these articles, known techniques, such as injection molding and extrusion molding, may be employed.

[ 0033]

### [ INVENTIVE EFFECTS ]

As detailed above, the resin composition based on crystalline polypropylene according to the present invention can be processed into articles exhibiting superior appearance without suffering from occurrence οf flow mark, while attaining superior mechanical properties, such as strength and so on, because the resin composition have contents of specific elastomer, specific crystalline polypropylene and specific talc each in a specific proportion range.

[ 0034]

[ MODE FOR EMBODYING THE INVENTION]

Below, the present invention will be described by way of Examples:

## EXAMPLES 1 to 10 and COMPARATIVE EXAMPLES 1 to 10

Preparation of the Resin Composition based on Crystalline Polypropylene

Using the elastomer as given in Table 1, the crystalline polypropylene as given in Table 2 and the talc given in Table 3, the resin compositions based on crystalline polypropylene as given in Tables 4 and 5 were prepared.

[ 0035]

Table 1

		E	astome	er	
	X-1	X-2	X-3	X-4	X-5
Content (wt. %) of					
Styrene	0	0	13	30	40
Ethylene	69	72	-	_	-
Propylene	29	25	-	-	-
5-ethylidene-2-norbornene	2	3	. <b>-</b>	_	
23℃ p-xylene-soluble component	100	100	100	100	100
Intrinsic viscosity (dl/g)	2.3	1.2	1.1	1.6	1.4
MFR (g/10 min.)	0.4	7.3	9	· 2	5
Polymer type	EPDM	EPDM	SEBS	SEBS	SEBS

[ 0036]

[Table 2]

Table 2

	Crys	stallir	ne poly	propy1	ene (F	PP)
	Y-1	Y-2	Y-3	Y-4	Y-5	Y-6
MFR (g/10 min.)	60	58	. 56	60	58	60
Ethylene content (wt. %)	0	0	0	0	3.0	0
23°C p-xylene- soluble component	<b>-</b> .	-	<b>-</b>	- :	EPR	-
Wt. % of 23 ℃ p- xylene-sol. comp.	0	0	0	0	11.5	0
Wt. % of 135℃ p- xylene-sol. comp.	100	100	100	100	100	100
mmmm (%) *1	98.6	98.5	98.7	98.4	98.1	96.4
Mw/Mn *2	11.0	25.5	6.0	37.1	7.6	11.3
Mz/Mw *3	10.2	9.3	3.6	5.7	10.6	10.5

[ 0037]

Notes \*1: Isotactic pentad proportion (mmmm) determined as the proportion of mmmm peak among the total absorption peaks in the methyl carbon region in the '3 C-NMR spectrum.

\*2: Conditions for the determination of numberaverage molecular weight (Mn), weight-average molecular weight (Mw) and z-average molecular weight (Mz) were as follows:

GPC : Model 150 C of the firm Waters.

Column: Model PL mixed B of the firm Polymer

Laboratories.

Amount of the sample = 400  $\mu$  1 with polymer

concentration of 0.15 % by weight.

Temperature : 135 ℃.

Solvent : o-dichlorobenzene

Mn, Mw ans Mz for each sample were determined under calibration onto each calibration curve prepared preliminarily using standard polystyrenes supplied from the firm Polymer Laboratories. For the analysis, an analysis software "Millenium" supplied from the firm Waters.

[ 0038] [ Table 3 ]

Table 3

	Tá	alc
	Z-1	Z-2
Average particle size $(\mu m)$	2.4	6.3

[ 0039]

[Table 4]

Table 4

Component (proportion in weight %)		I	Examp	ole				mpara ample		2
weight s)	1	2	3	4	5	6	1	2	3	4
Elastomers										
X-1	30	30	26	24	16	22	30	24	30	24
X-2	-	-	_	. –	-	11	-	-	-	-
x-3	-	_	_	_	8	_	-	-	_	-
X-4	-	-	_	-	-	-	-	-	-	- ·
X-5	-	_	-	-	-	-	_	_	_	_
Crystalline PP										
Y-1	50	_	27	56	56	51	_	_		-
Y-2	-	50	-	-	_	_	-	_	_	_
Y-3	-	_	-	-	-	-	50	-	-	56
Y-4	-	_	-	-	_	-	-	-	50	-
Y-5	_	-	27	-	-	-	_	56		_
Y-6	-	-	_	-	-	-	_	-	-	_
Talc										
Z-1	20	20	20	20	20	16	20	20	20	20
Z-2	-	-	_	_	-	_	_	_	_	-
Compnt.(a) *1	30	30	30	24	24	33	30	30	30	24
Compnt.(b) *2	50	50	50	56	56	51	50	50	50	56
Compnt.(c) *3	20	20	20	20	20	16	20	20	20	20

Notes \*1, \*2 and \*3: See under Table 5.

[ 0040]

[Table 5]

Table 5

Component (proportion in weight %)	F	Examp	ole				mpara mple	ative	2	-
weight o)	7	8	9	10	5	6	7	8	9	10
Elastomers										
X-1		-	-	_	-	_	-	· -	-	-
X-2	-	-	_	-	-	-	-	_	_	-
X-3	12	12	11	_	12	9	12	-	12	12
X-4	12	12	11	26	12	9	12	-	12	12
<b>x-</b> 5	-	-	-	-	-	-	-	24	_	- :
Crystalline PP										
Y-1	56	_	29	54	-	_	-	56	-	56
Y-2	-	56	-	-	_	-	-	-	_	_
Y-3	_	_	-	-	56	-	_	-	_	· –
Y-4	-	-	-	_	-	-	56	-	-	-
Y-5	-	-	29	-	_	62	-	-	-	_
Y-6	_	-	-	-	. –	-	-	-	56	-
Talc										
Z-1 .	20	20	20	20	20	20	20	20	20	-
Z-2	-	-	-	-	-	-	-	-	-	20
Compnt.(a) *'	24	24	24	26	24	24	24	24	24	24
Compnt.(b) *2	56	56	56	54	56	56	56	56	56	56
Compnt.(c) *3	20	20	20	20	20	20	20	20	20	20

[ 0041]

Notes for Tables 4 and 5:

\*2: The 135 $^{\circ}$  paraxylene-soluble and 23 $^{\circ}$  paraxylene-insoluble component (b).

\*3: The 135°C paraxylene-insoluble component (c). [ 0042]

2) Material Properties of the Resin Composition based on Crystalline Polypropylene

For the resin compositions based on crystalline polypropylene given in Tables 4 and 5, the material properties etc. were examined. The test specimens tests the were prepared by injection molding each composition on an injection molding machine (Model J100 SA II of The Japan Steel Works, Ltd.) at a cylinder set temperature of 230 °C and a metal mold 6 and 7.

[ 0043]

				·	
Features of the composi- tion	Component (C)	Component (B)	Component (A)		
MFR (g/10 min.) *3 FM (MPa) *4 IZ (J/m) *5 HR (Rockwell R) *6 BT (°C) *7 Flow mark occurrence rate (%) *8	Content (wt. %) Average particle size (µm)	Content (wt. %) mmmm (%)  Mw/Mn  *2  Mz/Mw  *2	Content (wt. %) Styrene cont. (wt. %) Intrinsic viscosity (d1/g)		
12 1850 650 47 -40.2	20 2.4	50 98.6 11.0 10.2	30 0 2.3	1	
11 1960 560 49 -37.2	20 2.4	50 98.5 25.5 9.3	30 0 2.3	2	
13 1830 600 48 -39.1 24	20 2.4	50 98.3 9.3 10.4	30 0 3.4	3	Example
14 2150 300 71 -20.5	20 2.4	56 98.6 11.0 10.2	24 0 2.3	4	ple
18 2170 400 71 -25.5	20 2.4	56 98.6 11.0	24 4 1.7	σı	
21 1720 615 50 -42.7 23	16 2.4	51 98.6 11.0 10.2	33 0 1.9	6	
12 1740 640 43 -40.3	20 2. 4	50 98.7 6.0 3.6	30 0 2.3		93
12 1630 660 40 -37.8 25	20 2.4	50 98.1 7.6 10.6	30 0 3.7	. 2	mparativ
10 1810 150 46 -2.3	20 2.4	50 98.4 37.1 5.7	30 0 2.3	3	Comparative Example
15 1960 290 66 -18,4	20 2. 4	56 98.7 6.0 3.6	24 0 2.3	4	e

Tal

[ 0044]

			7	Example 8	ple 9	10	ι	6 00	Comparative Example	e Exa	P <sub>T</sub>	umple 9
·	Component (A)	Content (wt. %) Styrene cont. (wt. %) Intrinsic viscosity (d1/g)	24 .22 1.3	24 22 1.3	24 19 1.8	26 30 1.6	24 22 1.3	24 15 2.9		24 22 1.3	24 24 22 40 1.3 1.4	
·	Component (B)	Content (wt. %) mmmmm (%)  Mw/Mn  *2  MZ/Mw  *2	56 98.6 11.0	56 98.4 25.5 9.3	56 98.3 9.3 10.4	54 98.6 11.0 10.2	56 98.7 6.0 3.8	56 98.1 7.6		56 98.4 37.1 5.7	56 98. 37. 5.	56 98.4 37.1 5.7
	Component (C)	Content (wt. %) Average particle size (µm)	20 2. 4	20 2.4	20 2.4	20 2.4	20 2.4	20 2. 4	20 2.4	0 20 .4 2.4		20 2.4
	Features of the composi- tion	MFR (g/10 min.) *3 FM (MPa) *4 IZ (J/m) *5 HR (Rockwell R) *6 BT (°C) *7 Flow mark occurrence rate (%) *8	29 2520 371 82 -32.3 28	27 2580 353 84 -30.1	30 2320 460 80 -31.1 24	28 2570 332 33 83 -30.2	31 2480 350 79 -32.8 89	33 2070 370 69 -25.2	33 2070 370 69 25.2 25	33 27 070 2630 150 370 150 39 83 5.2 3.5 27		27 2630 150 83 3.5 27

**\***∞

in ASTM D 746.

[ 0045]

Notes for Tables 6 and 7

- See Table 2
- See Table 2
- \*3: MFR was determined in Examples 1 6 and Comparative Examples 1 4 in accordance with the method prescribed in JIS K 7210 and, in Examples 7 - 10 and Comparative Examples 5 - 10, in accordance with that of ASTM D 1238.
- \*4: **\***5 FM (flexural modulus) was determined by the method prescribed in JIS K 7203 in Examples 1 - 6 and in Comparative Examples 5 - 10. Comparative Examples 1 - 4 as well as by the method prescribed in ASTM D 790 in Examples 7 - 10 and in (Izod impact strength) was determined in Examples 1 - 6 and Comparative Examples 1 - 4 Ş
- \*6: HR (surface hardness) was determined in Examples 1 - 6 and Comparative Examples 1 - 4 by the method prescribed in JIS K 7110 and in Examples 7 - 10 and Comparative Examples 5 - 10 by the method prescribed ASTM D 256, both at a temperature of 23°C. the method
- BT (brittle temperature) was determined in Examples 1 6 and Comparative Examples 1 4 by the method prescribed in JIS K 7216 and in Examples 7 - 10 and Comparative Examples 5 - 10 by the method prescribed prescribed in JIS K 7202 and in Examples 7 - 10 and Comparative Examples 5 - 10 by the method prescribed in ASTM D 685. In both methods. R was used for the steel sphere and evaluation was expressed in terms of
- Flow mark occurrence rate: flow mark is a regular pattern in stripes appearing on the face of molding in entire path length, which ratio was taken as the flow mark occurrence rate. of 3 mm, wherein the flow distance from the path inlet to the point at which a flow mark begins to occur effecting an injection molding of the composition on an injection molding machine using a metal mold molded article and belongs to a defect of appearance. the direction vertical to the resin flow due to imperfect transference of the mold face form onto the having an injection path of a spiral form having a length of 2.000 mm, a width of 10 mm and a thickness in order to determine the ratio of the path length within which the flow mark exists to the The flow mark occurrence rate was determined by

[ DESIGNATION OF THE DOCUMENT ] ABSTRACT

[ ABSTRACT]

### [ SUBJECT ]

To provide a resin composition based on crystalline polypropylene which can afford to produce molded articles exhibiting superior appearance with superior machanical material properties, such as strength etc., without causing occurrence of flow mark.

[ MEANS OF SOLUTION ] The resin composition based on crystalline polypropylene comprises

- (a) a component soluble in paraxylene of 23  $^{\circ}$ C, in an amount of 20 35  $^{\circ}$ by weight,
- (b) a component soluble in paraxylene of 135% and insoluble in paraxylene of 23%, in an amount of 43-65% by weight, and
- (c) a component insoluble in paraxylene of 135%, in an amount of 15 22 % by weight,

#### wherein

the component (a) is composed substantially of an elastomer (A) having a content of styrene or its derivative in the range of 0 - 35 % by weight and an intrinsic viscosity, determined in decalin at 135  $^{\circ}$ C, in the range of 0.1 - 5 dl/g,

the component (b) is composed substantially of a crystalline polypropylene (B) having an isotactic pentad proportion (mmmm) of 97 % or higher, an Mw/Mn of 9 or higher and an Mz/Mw of 8 or higher and

the component (c) is composed substantially of a talc (C) having an average particle size of 1 - 5  $\mu$  m. [SELECTIVE DRAWING] None